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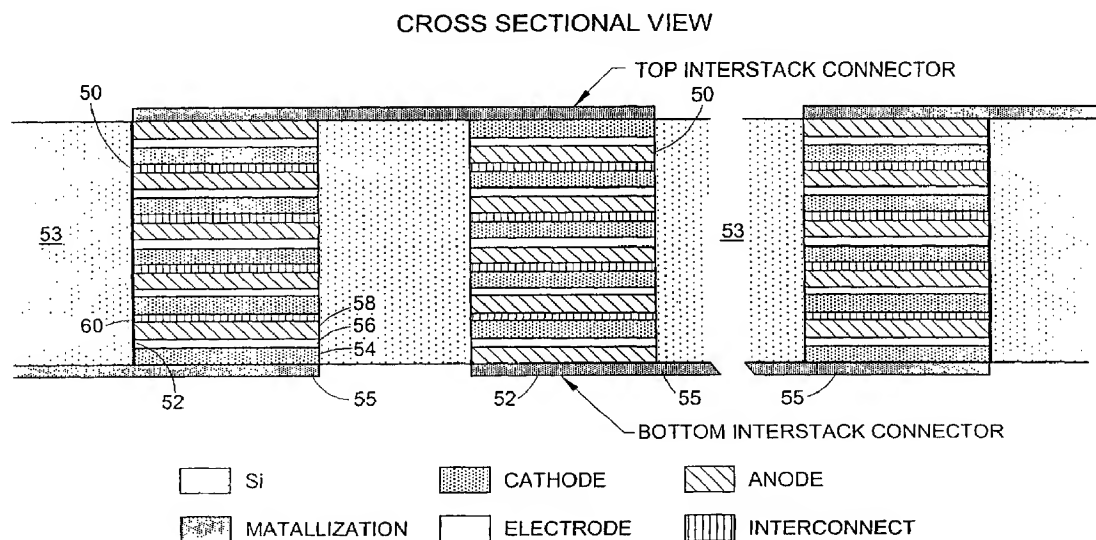
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(54) Title: **INK-JET BASED METHODOLOGIES FOR THE FABRICATION OF MICROBATTERIES**



(57) Abstract: A method of fabricating a microbattery (50) employs a microdispensing device (10) in the nature of an ink-jet printer to deposit a first electrode material (54), an electrolyte (56), and a second electrode material (58) in a stratified integral arrangement on a substrate or in a cavity (52). Separate ink-jet cartridges (30,32) are filled with the respective materials and appropriate adjustments are made to dispense appropriate amounts of droplets having predetermined rheological properties in a predetermined sequence to control the formation of electrical chemical battery components and the resulting microbattery (50).

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INK-JET BASED METHODOLOGIES FOR THE FABRICATION OF MICROBATTERIES

This application claims priority from U.S. Provisional Application Serial No. 60/197,974, filed April 14, 2000.

Background of the Invention

This invention pertains to the art of power supply, and more particularly to the art of electrochemical power supply useful in electronic and microelectromechanical devices. The invention is particularly applicable to a method of fabricating integrated microbatteries using ink-jet printing technology, and will be
5 described with particular reference thereto. It will be appreciated that the invention has broader applications evidenced in the flexibility associated with the microbatteries and their method of fabrication. For example, the method of fabrication lends itself to usefulness in the manufacturing of single layer flat batteries as well as batteries that can be stacked either inside pre-formed cavities or out.

10 Microelectromechanical systems (MEMS) technology provides for micron-sized complex engineering systems. MEMS integrated circuits combine logic circuitry with moving parts which enable interaction with other objects and systems. The MEMS require power to enable them to function, and such power is generally harnessed in the form of microbatteries. Existing microbatteries offer many positive attributes needed
15 for use in conjunction with MEMS and electronic devices. Such microbatteries, however, often require separate manufacture and installation and can sometimes lead to inefficiencies associated therewith.

Advances in the maturing field of ink jet printing have led to the development of procedures for the controlled and reproducible delivery of nanoliter
20 droplets of single phase liquids and particle dispersions. The small amounts of material contained in a single nanoliter droplet and the accuracy with which ejection can be

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performed enable creation of micrometer-sized three-dimensional stratified structures in complex two-dimensional patterns without the use of masks. Vertical structures of variable composition and thickness over very small lateral dimensions are also enabled. This is virtually unachievable by conventional lamination techniques.

5 Others have employed ink-jet printing techniques in connection with filling electrolyte material into existing or pre-fashioned screen or stencil printed electrodes and associated separator components. In such operations, a screen or stencil apparatus is used to first prepare an electrode and separator combination. Electrolyte is applied to the separator/electrode combination using an ink jet printer. A second electrode,
10 prepared by screen or stencil printing methodology, is then applied over the electrolyte-filled unit. Switching between the separate workpieces in preparing the cell is cumbersome and uneconomical and can lead to inaccuracies. This methodology requires the sequential employment of different apparatus and different printing technologies in completing the cell. Moreover, components fashioned by the stencil printing method are
15 limited in application. For example, it is not possible, or at least difficult, to form the microbatteries in a stacked formation inside a narrow cavity. Also, such microbatteries require the use of separation components adjacent the electrodes in order to prevent a short circuit.

There is a need to develop an integrated power source for
20 microelectromechanical systems and electronic devices which provide micron-sized engineering systems. An aim in this regard is to meet the required specifications for the operation of MEMS subdevices while maintaining a minimal overall volume. There is also a need for a method of microbattery fabrication that enables the assembly of electrode and electrolyte layers with a single printing apparatus such that the various
25 attributes of the layers can be controlled to enable the formation of complex lateral and vertical structures.

The present invention contemplates novel microbatteries and a novel method for fabricating such microbatteries that meets the aforementioned needs and others.

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Brief Description of the Invention

In accordance with the present invention, there is provided a novel method for fabricating microbatteries useful in microstructures such as MEMS devices.

In accordance with a more limited aspect of the invention, a method of
5 fabricating microbatteries calls for using a microdispensing device in the nature of an ink-jet printer to deposit a first electrode material, an electrolyte, and a second electrode material in a stratified integral arrangement on a substrate or in a cavity. Separate ink-jet cartridges are filled with the respective materials and appropriate adjustments are made to dispense droplets in a predetermined sequence to form microbatteries.

10 An advantage of the present invention is that the assembly of the entire microbattery is made with a single workpiece that quickly and efficiently forms a microbattery to precise specifications. The various attributes of the layers can be controlled to enable formation of complex lateral and vertical structures.

Another advantage of the present invention is that the method disclosed
15 herein enables the development of a microbattery that can be integrated directly into a microdevice and stacked as to increase voltage.

Another advantage is that the resulting battery does not require the use of separator elements. The electrolyte is configured to act as a separator and prevent the battery from shorting.

20 Yet, another advantage of the present invention is that the method enables the formation of a microbattery on any substrate such that it can be linked two dimensionally across the substrate.

Still other advantages and benefits of the invention will become apparent to those skilled in the art upon reading and understanding of the following detailed
25 description.

Brief Description of the Drawings

The invention may take physical form in certain parts and arrangements of parts, a preferred embodiment which will be described in detail in this specification
30 and illustrated in the accompanying drawings which form a part hereof.

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Figure 1 is a schematic diagram that exemplifies an ink jet printing device.

Figure 2A depicts the operation of filling a preexisting cavity with a stack of ink-jet printed batteries.

5 Figure 2B shows a resulting microbattery stack inside a cavity.

Figure 3 is a cross section schematic diagram of interconnected stacks of ink-jet fabricated cells in accordance with the present invention.

Figure 4 is a schematic diagram of an arrangement showing the electrochemical characterization of single ink-jet printed microbatteries.

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Detailed Description of the Preferred Embodiment

In accordance with the present invention, there is provided a novel method for the fabrication of on board integrated microbatteries for electronic and microelectromechanical devices. This technology relies on the sequential microdispersion or ink-jet
15 deposition of microdroplets of suspensions, slurries, dispersions, or solutions into pre-machined cavities to form the cathode, a polymer electrolyte, an anode and a metallic interconnect in a stratified-type arrangement. The microbatteries can be stacked into such cavities to yield high voltage subunits, which, in turn, can be connected either in series or in parallel to meet the demands of each and every MEMS subdevice or
20 electronic component. The technology also enables the deposition of microbatteries on complex two dimensional patterns without the need of masks, as well as fabrication of electrodes and electrolyte layers with graded or stratified structures along the normal to the axis of growth.

Ink-jet printing of batteries stems from Desktop Manufacturing (DM).
25 This technology enables the formation of complex three-dimensional structures via computer-aided design by adding material without the use of a mold. Ballistic Particle Manufacturing (BPM) is a DM approach that employs a computer controlled stream of particles to produce three-dimensional structures by printing sequential layers of materials, one on top of the other. Ink-jet injection (IJI), a class of BPM techniques,
30 provides a practical means of producing and controlling such arrays of particles.

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IJI technology has never been used in fabricating the multi-layer microbatteries of the present invention. Attention in this regard is directed to Figure 1 which sets forth a schematic diagram of an example of an ink-jet printing device 10. Carefully formulated inks are ejected as droplets 12 and in some cases charged by applying a voltage between the nozzle plate 14 and auxiliary electrode 16. The droplets can then be deflected by a high voltage applied to parallel deflector plates 18. Position of a substrate 20 is controlled by a computer. Appropriate synchronization enables the controlled build-up of a three dimensional structure. The properties of the droplets and the resulting structure are controlled by changing the composition and rheology of the inks. Rheological factors such as viscosity and surface tension, as well as nozzle shape and applied pressure, aid in controlling flow rate and drop formation to produce a desirable steady, satellite-free stream of droplets. Inks with viscosity in the range of 4.5 to 6.2 mPa and surface tension of around 22 mNm⁻¹ produce good deposits of particles of diameter about 0.1 to 0.2 μ m without excessive nozzle clogging.

Special "inks" are formulated. The inks comprise suspensions of oxides, carbon and metals that can be delivered using commercial ink-jet printer heads. Both single and stacks of batteries are produced using this methodology. The ability to stack the microbatteries enables a reduction in interconnect resistance. The ink-jet printing methodology performs at high rates without the need of expensive equipment. The cathodes, anodes and inks utilized in forming the integrated microbatteries and the ultramicrobatteries for MEMS and electronic devices are compatible with the operation of commercial ink-jet printer heads. A piezoelectric head/nozzle assembly 22 is used with fluids having desired or optimal rheological, hydrodynamic and other suspension fluid properties. The ink-jet printing of larger size batteries is also contemplated by this invention.

Sequential relative positioning between various ink-jet cartridges and the substrate enable the formation of the cathode, electrolyte, anode and interconnect in a stratified fashion to generate stacks capable of reaching voltage of several tens of volts. It is contemplated that this approach is similarly useful in producing electrodes with isotropic or graded compositions as well as to pattern batteries without the use of masks.

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Electrode/electrolyte/electrode assemblies are constructed by sequential deposition of electrode, electrolyte and electrode inks using different ink-jet heads to form single microbatteries. The same steps can be followed in stacking batteries in microcavities, as depicted in Figures 2A and 2B.

5 Figure 2A sets forth a schematic diagram of a battery stack formed into a cavity by ink-jet printing techniques. As will be noted, a series of cartridges 30, 32, 34, and 36 are aligned above a substrate 38. By providing relative motion horizontally between the substrate and the cartridges, a sequential deposition of layers via ink-jet printing technology is obtained. Relative movement between the substrate and cartridges
10 provides for sequential filling of cavity 40 in the substrate. The Figure shows in this instance the substrate moving to the right under the cartridges, though it is fully contemplated that the printing head move instead. The substrate shown in Figure 2A can be reposition in varying directions. Cartridge 30 represents the cathode material which has already been deposited into the substrate cavity 40. The substrate cavity is
15 shown under the electrolyte cartridge 32 which is depositing a microdroplet of the electrolyte into the cavity. Subsequently, the anode cartridge 34 will be aligned with the substrate cavity to form the anode layer followed by a deposition of the interconnect layer from the interconnect cartridge 36. The process is repeated by moving the substrate to reposition the cavity beneath the cathode cartridge 30 for the next sequential
20 laying of the subsequent battery. Figure 2B provides for the final schematic diagram of a battery stack 42 formed into a cavity by ink-jet printing techniques. Electrical connections 44 are shown above and below the stack.

Figure 3 shows a schematic diagram of arrays of interconnected battery stacks fabricated by the methodology of the subject invention. Layers of active material,
25 electrolyte, and interconnects are produced in a controlled and reliable fashion in terms of lateral and vertical directions. The batteries are shown as stacks 50 in cavities 52 formed in silicon 53. A cathode 54 is applied to a bottom interstack connect 55 via an ink-jet printing method. Electrolyte 56 is deposited on the cathode via the same ink-jet printing process. Thereafter, an anode 58 is deposited by ink-jet dispersion on the
30 electrolyte followed by the interconnect 60. The layers of cathode, electrolyte, anode,

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and interconnect are repeated several times, ending with the anode. The anode meets a top interstack connector **56** which then connects it to the next series of battery stacks.

Figure 4 shows a schematic diagram of an electrochemical characterization of a single ink-jet printed microbattery. A substrate **70** is provided.

5 Preferably, the substrate is patterned, though this is not required. The substrate may be formed by patterning metal layers onto smooth glass or silicon. A contact **72** is applied to the substrate via ink-jet printing followed by ink-jet printing of the anode **74**. An electrolyte **76** comprised of a polymer solution is applied next via ink-jet printing using independent ink-jet heads filled with electrolyte and electrodes, respectively. The
10 electrolyte **76** is depicted in an overhanging electrolyte geometry, such that contact is made with the substrate **70** and contact **72**. This overhanging geometry prevents potential electrical shorts between the electrode and the interface. As will be noted, the single microbattery of Figure 4 is assembled via ink-jet printing. A cathode **77** electrode is deposited on the electrolyte via ink-jet printing. Contact **78** leads from the cathode.

15 Preferably, the ink-jet fabrication of the single cathode, electrolyte and anode layers is conducted using a computer controlled Trident ink-jet head system which is available commercially. Inks useful in fabricating subject microbatteries are compatible with the nozzle through which they are developed. Preferably, the inks contain lithiated transitional metal oxides and carbon. The inks are preferably comprised
20 a dispersant such as an acrylic copolymer and a resin such as polyvinyl butyral, dissolved in an alcohol mixture, high area carbons and, if required, zirconia or alumina particles. Inks of similar rheological properties are formulated by incorporating materials for preparation of Li^+ electrodes. These include small particles of oxides, sulfides, or carbon as the active materials, high area carbon as a conductivity enhancer,
25 and a polymeric binder such as polyvinylidene fluoride (PVDF) and/or Li-ion conducting gels. Lithiated metal oxides of varying particles size are available from OMG Americas (Lakewood, Ohio) and, if needed, other commercial manufactures, or they can be prepared by known methods. High-area carbon and graphite particles can be obtained from Superior Graphite or other sources. Most nozzle dimensions on most commercial
30 ink-jet heads, including the Trident System, are on the order of only a few tens of μm ,

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making it necessary to reduce the size of the active particles well below 1 μm , which is smaller than those used in commercial Li-ion batteries. The amount of solid material delivered by individual droplets is small. Hence, hundreds of droplets are required to build up layers several micrometers thick.

5 Regarding electrolytes, in addition to using polyethylene oxide, gel electrolytes using polymethyl methacrylate (PMMA) as a basis, as well as advanced polymeric materials can be used. Alumina particles may be included in the polymer.

 The ink-jet printing technology is desirable because it provides for the integration of microbatteries directly onto microdevices. The high-energy-density, high-
10 power-density microbatteries that are capable of meeting the needs of onboard subunits involved in telecommunication, mechanical control, and other tasks are provided. Electrode and electrolyte layers are assembled with complex lateral and vertical structures in compositions with resolution on the order of micrometers. Such results would not be possible by other methods such as lamination. The microbatteries of the
15 present invention do not require the use of a separator between the electrode and the electrolyte. Indeed, the electrolyte provides the separator function. Note its overhanging position is Figure 4.

 The present invention is advantageous in that only a single type of technology, i.e., ink-jet printing, is required to form an entire battery or a series of
20 batteries. This enables the methodology employed to be more highly economical than the methodology employed with prior art microbatteries.

 Rechargeable, high-energy-density, high-power-density, lithium-ion polymer-electrolyte based micro-batteries can be assembled as single units or high voltage monolithic stacks according to the method of the present invention. Using the
25 novel ink-jet deposition strategy may have significant impact in achieving the full integration of power sources into microdevices. Examples could be microelectromechanical systems (MEMS) and labs-on-a-chip to meet the demands of a variety of on-board sub-systems, including motors, actuators, sensors, data storage and analysis, telecommunications, and other ancillary functions. The invention also concerns
30 the resulting ink-jet microbatteries.

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The procedures of the present invention meet the required specifications for the operation of MEMS subdevices, while minimizing the overall volume. Such subdevices include, but are not limited to, motors, sensors, data storage and analysis, transmission/reception units and other ancillary functions, such as actuators involved in
5 power and thermal management.

Although the procedure for fabrication of integrated microbatteries is of rather general applicability, the method provides, in particular, means for fabricating high energy density, high power density lithium-ion based microbatteries. These batteries incorporate lithium ion host lattices both for the anode and the cathode in the
10 form of micron size carbon and transition metal oxides or sulfides, respectively, and polymer based electrolytes including but not limited to polyethylene oxide and its derivatives or polymer based gelled electrolytes.

Miniaturization of lithium ion batteries is achieved by ink-jet printing techniques, in which each of the cell components (anode, electrolyte, cathode, and inter-
15 connect layers) is deposited sequentially to form the desired stratified structures as shown schematically in Figure 3. With the exception of the polymeric electrolyte, all other battery components consist of micron size particles, such as carbon for the anode, a transition metal oxide or sulfide as the cathode and a metal as the interconnect. Implementation of this methodology allows for stacks of batteries to be fabricated in
20 cavities micromachined into existing Si or SiC wafers or etched glass plates, which, by judicious interconnection, are able to supply the high voltages required for the operation of electrostatic motors, sensors, data storage and analysis, transmission/reception units, and other ancillary functions, such as actuators involved in power and thermal management and lab-on-a-chip applications. The on-board integrated microbatteries can
25 be interconnected in series or in parallel to meet the demands of virtually any MEMS subdevice or electronic component. The technology enables deposition of microbatteries on complex two dimensional patterns without the need of masks as well as fabrication of electrodes and electrolyte layers with graded or stratified structures along the normal to axis of growth.

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The invention has been described with reference to the preferred embodiment. Obviously modifications and alterations will occur to others upon a reading and understanding of this specification. It is intended to include all such modifications and alterations herein.

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Having thus described the preferred embodiment, the invention is now claimed to be:

1. A method of fabricating a microbattery, comprising the steps of:
depositing droplets of a first electrode material onto a substrate using a microdispensing device;
depositing droplets of an electrolyte material onto said deposited electrode
5 material using said microdispensing device to form an integral microstructure.
2. The method of claim 1, comprising the additional steps of:
depositing droplets of a second electrode material onto the microstructure using the microdispensing device; and
forming an integral microbattery structure.
3. The method of claim 1 wherein the microdispensing device is an ink-jet printer.
4. The method of claim 3, comprising the additional steps of:
filling a first ink-jet cartridge with a cathode material;
filling a second ink-jet cartridge with the electrolyte material;
filling a third ink-jet cartridge with an anode material;
5 sequentially dispensing droplets having suitable rheological properties from the first, second and third ink-jet cartridges to form a stratified microbattery structure.
5. The method of claim 4 wherein the substrate is a planar surface.
6. The method of claim 4 wherein the substrate defines a cavity therein into which the ink-jet cartridge materials are sequentially dispensed.
7. The method of claim 6 comprising the additional steps of:

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sequentially dispensing droplets of additional cathode, electrolyte and anode materials from the first, second and third ink-jet cartridges over the stratified microbattery structure to form stacked microbatteries for increases voltage.

8. The method of claim 7 comprising the additional step of:
forming a plurality of stacked microbatteries in a plurality of cavities defined in the substrate;
connecting the stacked microbatteries using a metallic interconnector.

9. The method of claim 8 comprising the additional step of:
dispensing droplets of the metallic interconnector from a fourth ink-jet cartridge filled with the metallic material.

10. A method of fabricating a microbattery, comprising the steps of:
depositing a first electrode onto a substrate using an ink-jet printer:
depositing an electrolyte onto the electrode using an ink-jet printer;
5 depositing a second electrode onto said electrolyte using an ink-jet printer to fabricate an integrated electrochemical cell.

11. The method of claim 10, including the additional step of:
controlling a microdispersion of particles deposited by the ink-jet printer.

12. The method of claim 10, including the additional step of:
functioning the electrolyte as a separator as well as electrolyte.

13. The method of claim 10, including the additional step of:
sequentially depositing a first electrode, an electrolyte and a second electrode on the microbattery of claim 1 to form a stacked microbattery arrangement.

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14. The method of claim 10, wherein the steps of depositing occur in a cavity.
15. The method of claim 10, wherein the steps of depositing occur in a linear arrangement.
16. A microbattery formed according to the method of claim 1.
17. The method of claim 10, including the additional step of:
depositing an interconnect on the second electrode to interconnect with another using a connector.
18. A microbattery comprising:
an anode, electrolyte, and cathode all formed by an ink-jet printing process such that the cathode and anode electrolyte are integrated as a single unit.

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FIG. 1

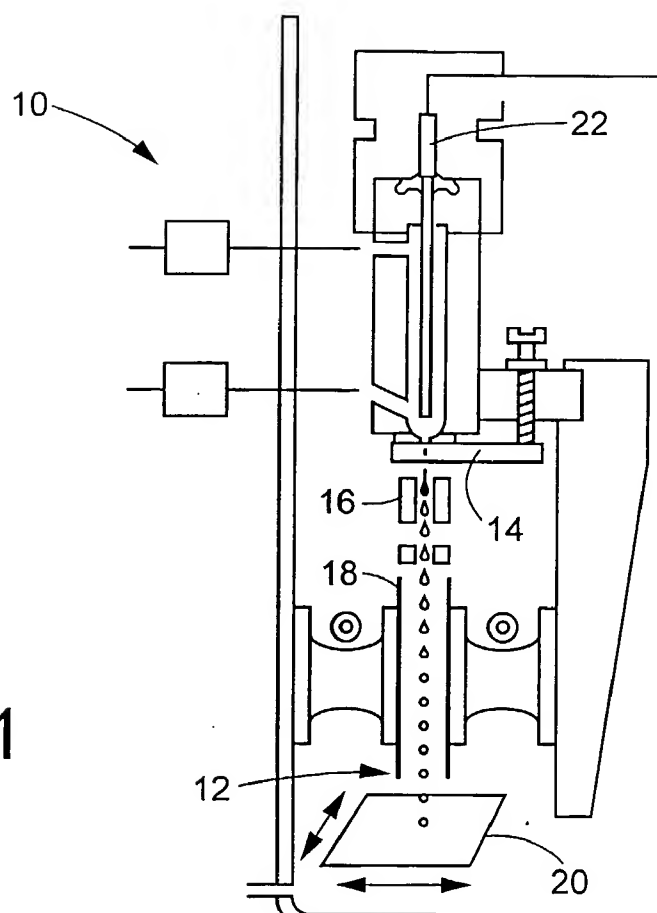
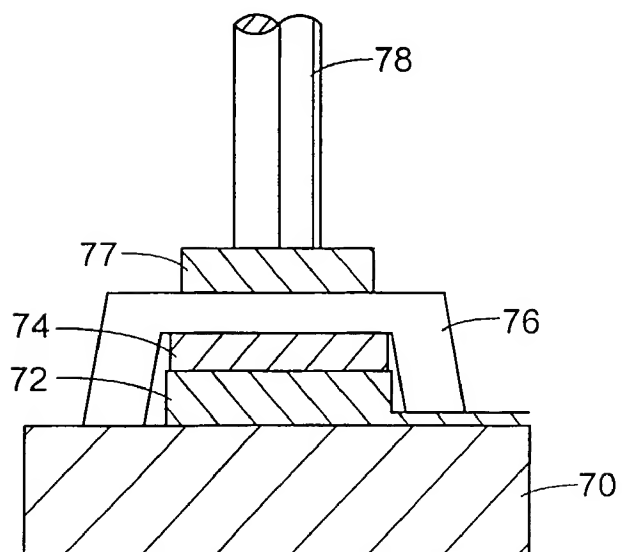
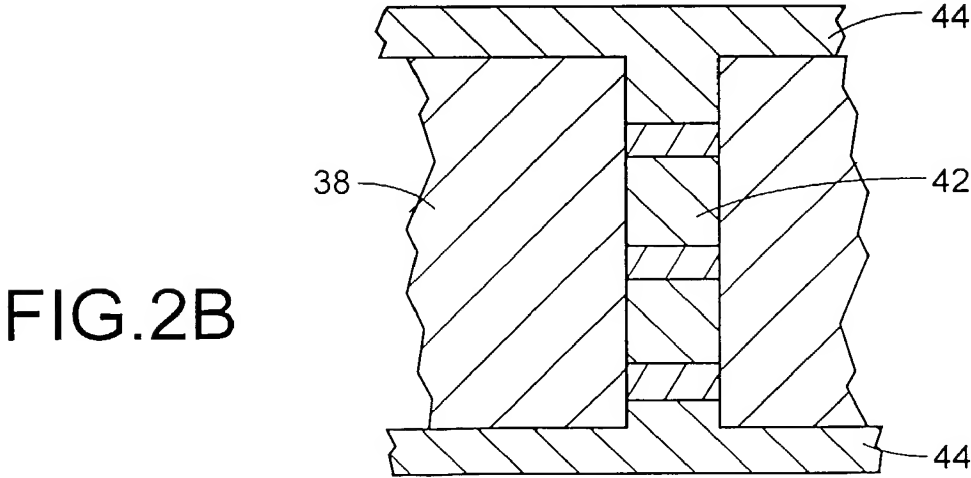
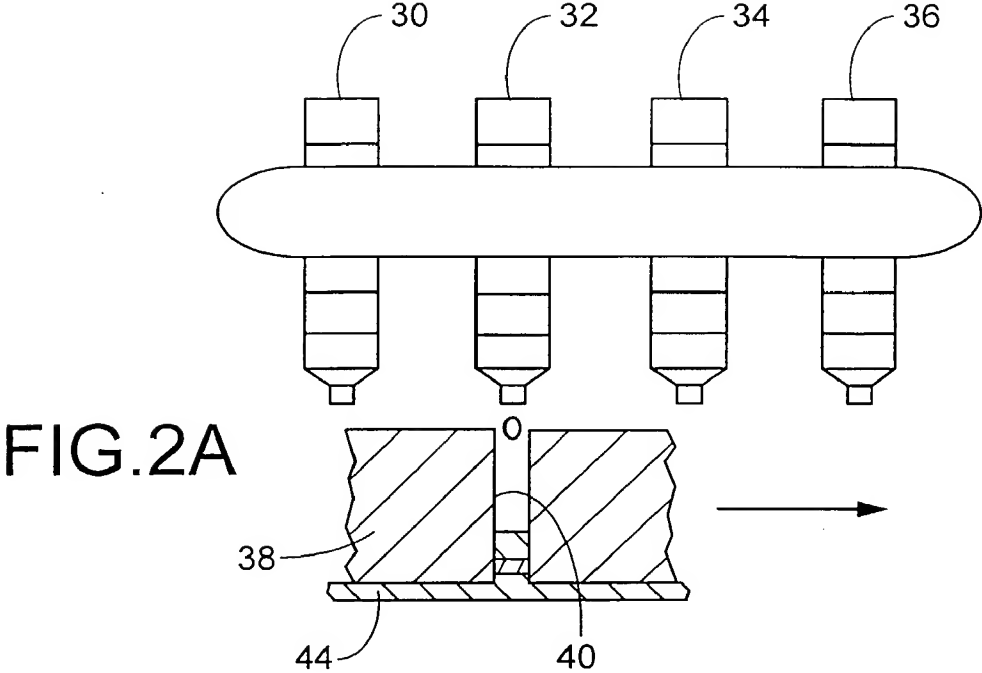


FIG. 4





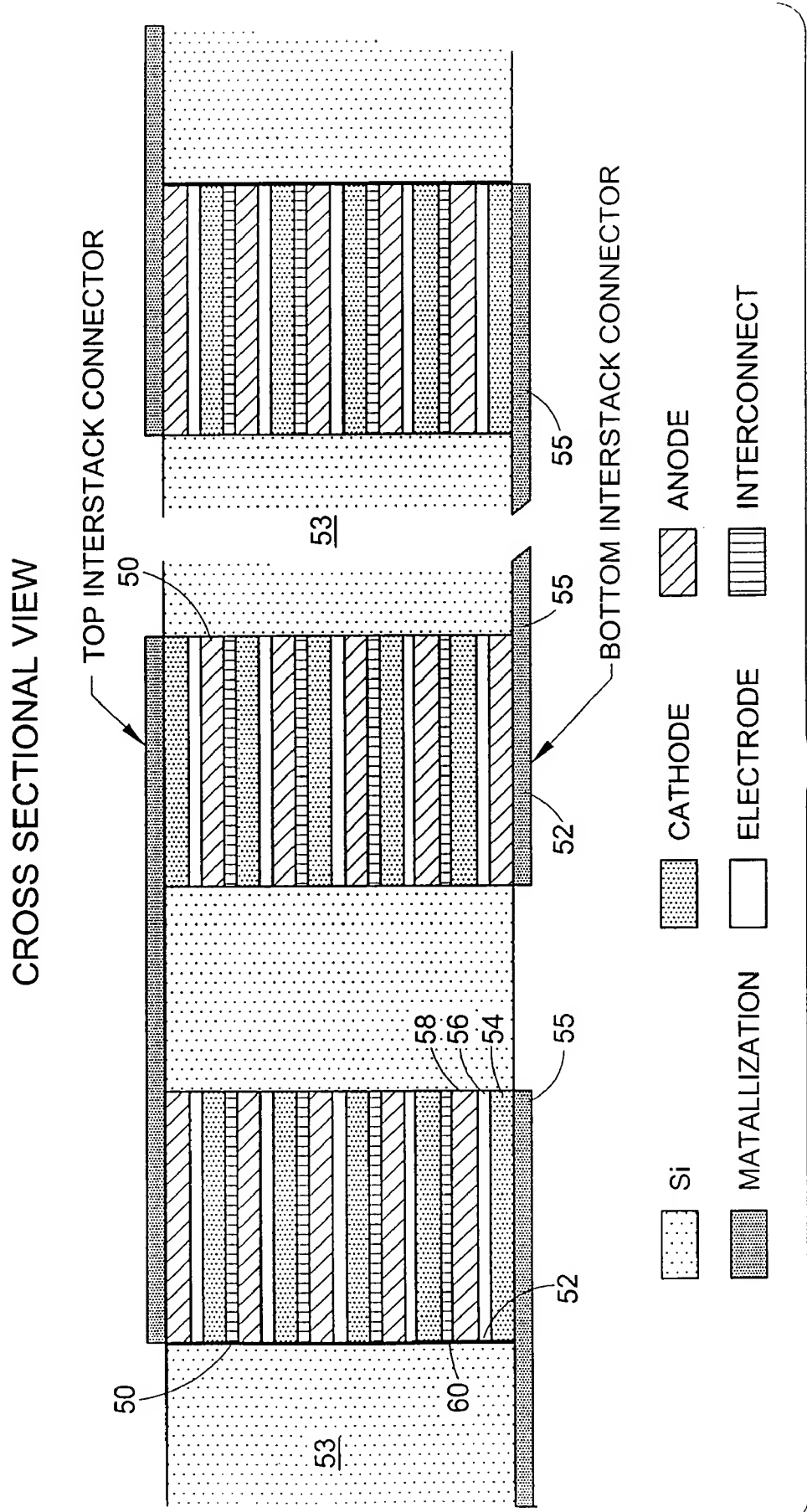


FIG. 3

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US01/12278

A. CLASSIFICATION OF SUBJECT MATTER

IPC(7) :H01M 6/18; 6/00
US CL :29/623.5

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 29/623.5, 623.1; 429/162, 127, 124

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched
NONE

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
Please See Extra Sheet.

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 3,375,135 A (MOULTON ET AL.) 26 March 1968, col. 3, line 74-col. 4, line 57, Fig. 2.	16, 18
X	US 5,338,625 A (BATES ET AL.) 16 August 1994, see col. 3, line 21-col. 4, line 8, Fig 3.	16, 18
X	US 5,705,293 A (HOBSON) 6 January 1998, see col. 3, line 42-col. 4, line 52, Fig 2.	16, 18
A	US 5,865,860 A (DELNICK) 2 February 1999	1-18
A	US 4,892,795 A (FANG ET AL.) 9 January 1990	1-18
A, P	US 6,117,593 A (STACHOVIK ET AL) 12 September 2000	1-18

☐ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

* Special categories of cited documents:	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
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B. FIELDS SEARCHED

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search terms: battery, batteries, ink jet, electrode, electrolyte, drop, droplet, liquid, micro,
microbattery, miniature